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Temporal trends in the levels of metals, PCDD/Fs and PCBs in the vicinity of a municipal solid waste incinerator. Preliminary assessment of human health risks

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ABSTRACT

The concentrations of PCDD/Fs, PCBs, and metals were determined in soil and air samples collected near a municipal solid waste incinerator (MSWI) located in Mataró (Catalonia, Spain), being the health risks for the population living in the vicinity of the facility subsequently assessed. We also established the temporal trends with respect to previous surveys performed in the same zone. In general terms, the highest environmental levels of the pollutants were found in the city of Mataró and were independent on the distance to the MSWI. Soil levels of metals did not show temporal variations between the current and previous campaigns. However, the concentrations of metals in air samples collected in 2011 and 2013, were lower than those found in our 2008 survey. Regarding PCDD/Fs and PCBs, no differences were noted between the levels of the current survey and those corresponding to 2008. Anyhow, the concentrations of metals, PCDD/Fs, and PCBs in soils and air did not exceed the reference values established by regulatory organizations, being also in the low range of other similar studies recently reported. Finally, the human non-carcinogenic and carcinogenic risks due to exposure to metals, PCDD/Fs, and PCBs, were, for the population living in the neighborhood of the MSWI, considered as acceptable according to international standards.

1. Introduction

In modern societies, the production of a continuous increasing volume of wastes has given place to the need to find more complex solutions as suitable treatment methodologies. According to data from the EC, Europe is using 16 tons of material per person and year, becoming 6 of them waste (EC, 2015). Although waste management has substantially improved in the EU, the European economy is still losing a significant amount of potential secondary raw materials, including metals, wood, glass, paper, plastics, etc. Early the current decade, total annual waste production in the EU amounted 2.5 billion tons, being only a limited share (36%) recycled. The rest was landfilled or burned, of which some 600 million tons could be recycled or reused (EC, 2015).

The Directive 2008/98/EC (EC, 2008a) sets the basic concepts and definitions related to waste management, such as definitions of waste, recycling, and recovery. It also highlights that waste should be managed without endangering human health and harming the environment, particularly preventing any risk to water, air, soil, plants or animals, any nuisance through noise or odors, and any impact on the countryside or places of special interest. The EU's approach to waste management is based on the "waste hierarchy", which sets the following priority order when shaping waste policy and managing waste at the operational level: prevention, reuse, recycling, recovery and, as the least preferred option, disposal, which includes landfilling and incineration without energy recovery.

In recent years, waste incineration has become a serious option of waste management in developed countries. Although this practice presents numerous advantages, such as energy recovery and volume minimization, public controversy is also frequent at those locations where municipal solid waste incinerators (MSWIs) are operating or being planned. Governments and health authorities are under increasing pressure from the public to prove the absence of potential adverse health effects produced by these activities, as well as other waste management practices (Giusti, 2009). MSWIs have been historically associated to emissions of toxic chemicals, such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and heavy metals (Cheng and Hu, 2010; Ragazzi et al., 2013; Schuhmacher et al., 2006; Vehlow, 2015), among other pollutants.

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In 2008, we initiated a wide environmental monitoring program around the MSWI of Mataró (Catalonia, Spain) in order to assess the potential health risks for the population living in the surroundings, associated to exposure to metals, PCDD/Fs and polychlorinated biphenyls (PCBs) (Rovira et al., 2010). The environmental levels of these pollutants were rather low compared with those found in other areas, not meaning the MSWI of Mataró a significant source of heavy metals, PCDD/Fs, and PCBs in the area (Rovira et al., 2010). In parallel, a number of biomonitoring studies on the concentrations of PCDD/Fs and PCBs in blood samples of the population of Mataró have been also performed between 1995 and 2012 (Gonzalez et al., 1998, 2000; Parera et al., 2013).

In July 2010, the new Integrated Waste Recovery Centre (IWRC) of Maresme located in Mataró was inaugurated. This infrastructure includes a new plant for the treatment of refuse waste fraction, with a capacity of 190,000 tons/year to recover all usable materials contained, feeding the MSWI with non-usable materials. Additionally, several improvements were made to adapt the existing lines of the MSWI to these new refuse derived fuel (Rada and Andreottola, 2012). The two lines of the MSWI have a capacity of 160,000 tons/year. In addition, the IWRC of Maresme also owns waste transfer stations especially allocated for the organic fraction of municipal solid waste (biowaste), light packaging, paper/cardboard, and glass (CTRSUM, 2015a,b). Air pollution control systems of Mataró MSWI include fabric filters (PTFE + P84), selective (SCR) and non-catalytic reduction (SNCR) for NO_x abatement, and semi-dry Ca-based scrubber, before the exhaust air emission through a 45 m-high stack (CTRSUM, 2015b).

The objective of the present study was to update the information regarding the environmental burdens of PCDD/Fs, PCBs and metals in the vicinity of the IWRC of Mataró, after the renewal of the facility. The temporal trends of the concentrations of the analyzed pollutants in air and soil were established, while human health risks for the population living in the vicinity of the new Integrated Waste Recovery Centre of Mataró, were also assessed.

2. Materials and methods

2.1. Sampling

The IWRC of Maresme is placed in Mataró, a 124,000 inhabitants city located at 25 km NE from Barcelona. In November of 2011, and again in November of 2013, we collected four air and eight soil samples at various points around the MSWI. Four air and 4 soil samples were collected in Mataró city (under the premises of the facility (MSWI), at <1 km (NE1), and at >1 km (NE2)), and one in the village of Arenys de Mar (12 km from the facility), which was used as a background point (B). The four air samples were collected in the same week, being two of them

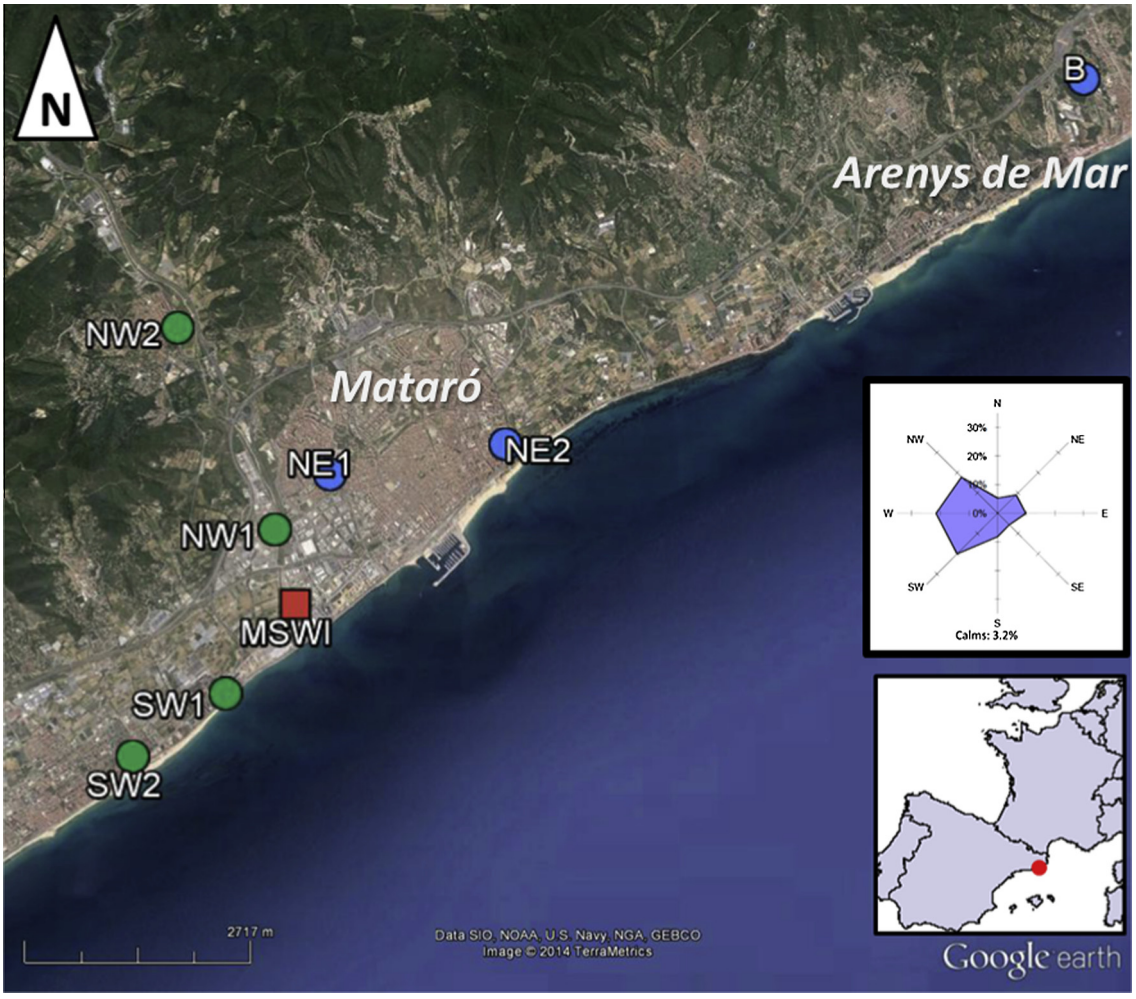

Fig. 1. Soil sampling points marked in green, those three points in blue are air and soil sampling points. In red, MSWI location and air and soil sampling point. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

obtained from Monday to Wednesday, and the other two from Wednesday to Friday. These sampling points were located at the same points than those previously reported (Rovira et al., 2010). Four additional soil samples were also collected as follows: two samples (at different distances (<1 km and >1 km)) in both directions regarding the facility: south-west (SW1 and SW2) and north-west (NW1 and NW2). The eight soil samples were collected during the same day. The locations of the eight soil and four air sampling points are detailed in Fig. 1. Mean temperatures during the samplings were 14 °C (range from 8 °C to 21 °C) and 8 °C (range from 3 °C to 16 °C) in November 2011 and November 2013, respectively. Accumulated precipitations were 99 and 115 mm in November 2011 and November 2013, respectively. There was no rainfall during the sampling. Prevalent winds in the area come from north, northeast, and southwest (Fig. 1).

Soil and air sampling procedures were previously described (Rovira et al., 2010). Briefly, five soil subsamples in an area of around 25 m^2 were collected from the surface layer (0–5 cm depth). Soils were kept in polythene bags and dried at room temperature. Once dried, soil samples were sieved through a 2 mm mesh screen to standardize particle size. Air sampling was conducted following the European directive 2008/50/EC on air quality (EC, 2008b). For the sampling of PCDD/Fs and PCBs, high-volume active samplers TE-1000 (Tisch Environmental, Cleves, OH, USA) were used, according to the US EPA method TO-9. The particle phase of PCDD/Fs and PCBs were collected on quartz fiber filters (QFFs), while the gas phase was collected by means of polyurethane foams (PUFs). In order to remove any impurity, PUFs were pre-cleaned with dichloromethane (Scharlau, Sentmenat, Spain) in a Soxhlet equipment for 24 h. Air volumes for the sampling of PCDD/Fs and PCBs were ranged between 520 and 750 m^3, lasting 48 h. QFFs and PUFs were stored together in amber glass bottles, and frozen at −20 °C until analysis. For the sampling of heavy metals associated with PM_{10}, we used a high-volume active sampler DV-6070 (Tisch Environmental, Cleves, OH, USA), according to the US EPA method IO-2.1. Sampling duration took around 24 h, collecting a volume of air ranging 1700–2100 m^3. After the sampling, QFFs were properly put in a cool dry place until the analysis.

2.2. Analytical procedure

2.2.1. Organochlorinated compounds

The analyses of PCDD/Fs and PCBs in soil samples were done by High Resolution Gas Chromatography coupled to High Resolution Mass Spectrometry (HRGC/HRMS), in combination with the isotope dilution technique, according to the US EPA method 1613 for PCDD/Fs, and a combination of the US EPA Method 1668 and JIS K 0311, for PCBs. The concentrations of PCDD/Fs and PCBs in air were also determined by HRGC/HRMS, following the German VDI 3499 method. To control the whole process and to evaluate potential losses, labelled extraction standards ($^{13}C_{12}$-PCDD/Fs and $^{13}C_{12}$-PCB congeners) were added. An Accelerated Solvent Extraction (ASE) was carried out by using toluene. The clean-up procedure was carried out by adsorption chromatography on a mixed silica column and adsorption/fractionation on alumina. Recovery percentages of PCDD/Fs ranged between 32% and 130% and between 67% and 140%, for soil and air samples, respectively. The limits of detection (LODs) of the 17 PCDD/F congeners in air were: 0.01 pg/m^3 for tetra-, penta-, and hexa-substituted congeners, and 0.07 pg/m^3 for hepta- and octa-substituted congeners. The LODs of the 17 PCDD/F congeners in soils were the following: 0.04 ng/kg for 2,3,7,8-TCDD, 0.25 ng/kg for 2,3,7,8-TCDF, 0.08 ng/kg for penta- and hexa-substituted congeners, and 0.18 ng/kg for hepta- and octa-substituted congeners. The LODs for the PCBs ranged between 32 and 36 ng/kg for soils, being 0.001 ng/m^3 for air samples. Total concentrations of PCDD/Fs were calculated by using the toxic equivalency factors (TEFs) of the World Health Organization (WHO-TEFs) (van den Berg et al., 2006).

2.2.2. Metals

Soil samples (0.5 g) were digested with 5 mL of HNO_3 (65% Suprapur, E. Merck, Darmstadt, Germany) in a Milestone Start D Microwave Digestion System, for 10 min until reaching 165 °C, and kept at this temperature for 20 min. On the other hand, a portion (50 cm^2) of QFFs was treated with a mixture of 2 mL of HNO_3 (65% Suprapur, E. Merck, Darmstadt, Germany) and 3 mL of HF (37.5%, Panreac, Castellar del Vallès, Spain) in hermetic Teflon bombs for 8 h at room temperature and 8 h at 80 °C. The extracts were subsequently evaporated in a sand bath, and reconstituted with HNO_3. The concentrations of arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), antimony (Sb), tin (Sn), thallium (Tl), vanadium (V), and zinc (Zn) were determined by means of inductively coupled plasma mass spectrometry (ICP-MS). Blank and control samples, as well as reference materials (Soil, Loamy clay, Resource Technology Corporation US), were used to check the accuracy of the instrumental methods. The recovery percentages of the reference material ranged between 87% and 107% and between 67% and 167%, in soil and air samples, respectively. The detection limits in air were: 0.22 ng/m^3 for As, Cu, Hg, Ni, and Sb; 0.54 ng/m^3 for Cr, V, and Zn; 0.11 ng/m^3 for Co and Sn; and 0.05 ng/m^3 for Cd, Mn, Pb, and Tl. In soils, the LODs were the following: 0.25 mg/kg for Cr, V, and Zn; 0.10 mg/kg for As, Cu, Hg, Ni, and Sb; 0.05 mg/kg for Co and Sn; and 0.03 mg/kg for Cd, Mn, Pb and Tl.

2.3. Human health risks

The calculations of the exposure through ingestion, dermal absorption and inhalation, as well the estimation of the associated health risks, were done as previously described (Rovira et al., 2010). The numerical expressions were taken from the Technical Guide of the "Spanish Royal Decree 9/2005" (Ministerio de la Presidencia, 2005), which establishes the list of activities potentially contaminants of soils, as well as the criteria and standards for the declaration of contaminated soils (Ministerio de Medio Ambiente, 2007). It is based on the US EPA RAGS methodology (US EPA, 1989). In addition, inhalation risks were calculated based on the most updated US EPA RAGS methodology for this route of exposure (US EPA, 2009a). Toxicological data were obtained from the Risk Assessment Information System database (RAIS, 2013), with the exception of the oral reference dose for Pb, which was taken from the WHO (Seiler et al., 1988). To assess the carcinogenic and non-carcinogenic risk from dermal exposure, the oral reference dose (RfDo) and the oral slope factor (SFo) were multiplied and divided, respectively, by the gastrointestinal absorption factor (GI_{ABS}) (US EPA, 1989). Specific equations of human exposure and risk characterization have been provided by Rovira et al. (2010).

2.4. Statistics

For data analysis, substances below limits of detection (LODs) were assumed to have a concentration equal to one-half of the respective LOD (ND = ½ LOD). Data analysis was carried out by means of the statistical software package SPSS 20.0. The level of significance was set at a probability lower than 0.05 ($p < 0.05$). To evaluate significant differences between groups, the Levene test was applied to verify the equality of variances. ANOVA or Kruskal Wallis tests were subsequently applied depending on whether the data followed a normal distribution, or not, respectively.

3. Results and discussion

3.1. Concentrations in soils

Table 1 summarizes the levels of metals, PCDD/Fs and PCB in soils inside the MSWI, collected at different distances (<1 km and >1 km) of the facility, as well as those at the background (B) sampling point. In general terms, the highest levels were detected in the sampling points of Mataró (<1 km and >1 km). The only exceptions were various metals (Co, Cr, Mn, Ni, Sn, Tl, V, and Zn) which showed higher levels at the background area. For all analyzed trace elements and organic compounds, no significant differences were noted in their levels, according to the distance to the plant ($p > 0.05$), in both sampling campaigns, 2011 and 2013. Taking into account the wind directions, significant differences ($p < 0.05$) were found in the PCDD/F levels between the northwest (NW) (0.44 ng WHO-TEQ/kg) and southwest (SW) (0.13 ng WHO-TEQ/kg) directions, whereas significantly ($p < 0.05$) higher PCB levels were found in the NW direction (3276 ng/kg). With respect to the temporal trends, no significant ($p < 0.05$) differences were observed for the concentrations of metals, PCDD/Fs and PCBs in soils between the 2011 and 2013 campaigns. When comparing with the results of the background survey, performed in 2008, a significant ($p < 0.05$) reduction of the mean levels of As between 2008 (1.96 mg/kg) and 2011 (0.78 mg/kg), and of Cd between 2008 (0.14 mg/kg) and 2013 (0.05 mg/kg), was noted. In turn, none of the remaining target chemicals presented significant changes of concentration in soil, a long-term environmental monitor. The congener profiles of PCDD/Fs and PCBs in the three campaigns (2008, 2011, and 2013) are depicted in Figs. S1 and S2 (Supporting Information), respectively. PCDD/Fs showed similar profiles in the two last campaigns 2011 and 2013, with lower contributions (68%) of OCDD than in the 2008 campaign (78%) (Rovira et al., 2010). The soil PCB congener presented different profiles in each sampling campaign (2008, 2011, and 2013) (Fig. S2). The comparison of these results with threshold values for metals and metalloids in soils, set by the Agency of Waste of Catalonia (ARC, 2009), shows that none of the elements exceeded the most restrictive levels, established for non-urban or non-industrial soil uses.

Table 1
Mean concentrations of metals (mg/kg), PCDD/Fs (ng WHO-TEQ/kg), and PCBs (ng/kg) in soil samples collected inside and in the vicinity of the MSWI of Mataró, and in a control point (B).

	2011				2013			
	MSWI $n = 1$	<1 km $n = 3$	>1 km $n = 3$	B $n = 1$	MSWI $n = 1$	<1 km $n = 3$	>1 km $n = 3$	B $n = 1$
As	0.65	0.87	0.72	0.97	1.14	1.12	1.85	0.73
Cd	ND	0.05	0.09	0.05	ND	0.06	0.05	0.03
Co	1.32	5.80	4.25	10.4	2.73	5.78	5.00	9.10
Cr	2.62	10.3	6.91	17.1	4.95	9.99	8.21	13.6
Cu	3.19	10.4	12.3	8.29	5.67	19.4	14.2	8.69
Hg	ND	ND	ND	ND	ND	ND	ND	ND
Mn	96.5	350	336	754	152	360	366	683
Ni	1.15	2.75	1.91	4.17	2.54	3.43	2.76	3.71
Pb	3.18	9.41	8.77	11.5	5.10	11.6	20.4	10.3
Sb	ND	ND	ND	ND	ND	ND	ND	ND
Sn	0.25	0.69	0.81	1.34	0.49	0.98	1.01	1.48
Tl	0.04	0.19	0.15	0.30	0.09	0.18	0.18	0.30
V	9.57	55.0	37.8	95.5	19.2	61.2	45.5	93.4
Zn	17.1	56.5	50.0	90.5	22.4	62.0	55.9	77.4
PCDD/Fs	0.13	0.28	0.24	0.13	0.16	0.43	0.39	0.12
PCBs	ND	1823	293	750	406	2593	1732	46.0

ND: Not detected.
<1 km: NW1, SW1, and NE1.
>1 km: NW2, SW2, and NE2.
PCBs: Sum of the 7 environmental indicator congeners: PCB 28, 52, 101, 118, 153, 138, and 180.

Table 2
Summary of some recent studies reporting levels of PCDD/Fs and PCBs in soil samples, including the results of the current study.

PCDD/Fs (ng TEQ/kg)	Type	Location	References
Levels around a MSWI			
0.47–2.07	Range	Tianjin, China	Liu et al. (2013b)
0.58 (0.11–1.35)	Mean (range)	Tarragona, Spain	Vilavert et al. (2012)
1.73–2.06	Mean	Taiwan	Wang et al. (2013)
2.00	Mean	Taiwan	Chen et al. (2011)
1.35 (0.57–2.0)	Mean (range)	Rimini, Italy	Vassura et al. (2011)
4.73 (0.64–61.15)	Mean (range)	Shanghai, China	Deng et al. (2011)
0.34 (0.14–0.46)	Mean (range)	Mataró (Spain 2008)	Rovira et al. (2010)
0.23 (0.13–0.56)	Mean (range)	Mataró (Spain 2011)	Present study
0.34 (0.12–0.61)	Mean (range)	Mataró (Spain 2013)	Present study
Other studies			
6.8 (3.6–15.7)	Mean (range)	Parks Moscow (Russia)	Shelepchikov et al. (2011)
5.6 (0.27–15.4)	Mean (range)	Urban Moscow (Russia)	Shelepchikov et al. (2011)
5.7 (0.67–16.1)	Mean (range)	Industrial Moscow (Russia)	Shelepchikov et al. (2011)
0.06–49.3	Range	Republic of Maldives (Developed)	Colombo et al. (2014)
0.02–15.8	Range	Republic of Maldives (Undeveloped)	Colombo et al. (2014)
2.3 (1.7–3.2)	Mean (range)	Parks Beijing (China)	Li et al. (2014)
0.19–1.05	Range	Taurus Mountain (Turkey)	Turgut et al. (2012)
1.09/2.54	Mean	Zhejiang (China 2007/2010)	Li et al. (2012)
PCBs (ng/kg)	**Type**	**Location**	**References**
Levels around a MSWI			
28.0–264.4	Range	Tianjin, China	Liu et al. (2013a)
27140	Mean	Guangzhou (China) impacted	Zeng et al. (2014)
5270	Mean	Guangzhou (China) control	Zeng et al. (2014)
1446 (167–3340)	Mean (range)	Mataró (Spain 2008)	Rovira et al. (2010)
888 (ND-5000)	Mean (range)	Mataró (Spain 2011)	Present study
1679 (46–5909)	Mean (range)	Mataró (Spain 2013)	Present study
Other studies			
5921	Mean	Tarragona, Spain (Industrial)	Nadal et al. (2011)
2773	Mean	Tarragona, Spain (Urban)	Nadal et al. (2011)
605	Mean	Tarragona, Spain (Unpolluted)	Nadal et al. (2011)
19–1321	Range	Brusa (Turkey)	Salihoglu et al. (2011)
3540 (ND-11460)	Mean (range)	Beijing (China)	Wu et al. (2011a)
11,400 (2800–40500)	Mean (range)	Parks Moscow (Russia)	Shelepchikov et al. (2011)
10,000 (2100–32,200)	Mean (range)	Urban Moscow (Russia)	Shelepchikov et al. (2011)
14,800 (3900–50,800)	Mean (range)	Industrial Moscow (Russia)	Shelepchikov et al. (2011)
70.8–228	Range	Taurus Mountain (Turkey)	Turgut et al. (2012)

Various countries have already established benchmarks (US EPA, 2009b) with respect to PCDD/Fs in soils. Thus, Austria, Sweden and Finland have taken a maximum acceptable concentration of 10 ng I-TEQ/kg for all land uses, while Canada has reduced this limit to 4 ng I-TEQ/kg (US EPA, 2009b).

These maximum admissible levels were not reached in the present study. Table 2 summarizes PCB and PCDD/F levels in samples of surface soils, reported in recently published studies over the world. Both chemicals showed values generally lower than those reported in those studies.

3.2. Concentrations in air

Table 3 summarizes the concentrations of metals associated to PM_{10}, PCDD/Fs, and PCBs in samples collected in the four air sampling points: one, under the premises of the MSWI, two in Mataró at different distances (<1 km and >1 km) of the facility, and the last one, considered as background. Clear trends for the concentration of metals, PCDD/Fs and PCBs according to the distance to the MSWI, were not detected. However, in general terms, the points in Mataró (<1 km and >1 km) showed the highest pollutant levels in air. It must be highlighted that the levels of Pb in 2011 (460 ng/m^3 at <1 km and 520 ng/m^3 at >1 km) and those of Cu in 2013 (132 ng/m^3 at <1 km and 103 ng/m^3 at >1 km) detected in the Mataró sampling points were particularly high. In relation to the temporal variability, mean levels of most of the analyzed elements (As, Cd, Co, Cr, Mn, Ni, Sb, Sn, and V) were significantly higher ($p < 0.05$) in 2008 than in the current campaigns (2011 and 2013). In turn, no significant differences with time were observed for the mean levels of PCDD/Fs or PCBs. The congener profiles of the collected air samples and those from the stack emissions are depicted in Fig. S3 (Supporting information). Stack emissions higher contribution for 2,3,7,8-TCDF congener and lower contributions for OCDD, 1,2,3,4,6,7,8,9-HpCDF, and OCDF congeners were found in both (2011 and 2013) sampling campaigns. The PCB congener contributions to the total PCB level are shown in Fig. S4 (Supporting information).

Table 3
Concentrations of metals (ng/m^3), PCDD/Fs (pg WHO-TEQ/m^3), and PCBs (ng/m^3) in air samples collected inside and in the vicinity of the MSWI of Mataró, and in a control point (B).

	2011				2013			
	MSWI	<1 km	>1 km	B	MSWI	<1 km	>1 km	B
As	0.76	0.78	0.78	0.51	0.46	0.55	0.24	0.32
Cd	0.20	0.31	0.45	0.11	0.24	0.22	0.21	0.10
Co	0.09	0.15	ND	0.05	0.11	0.18	0.12	ND
Cr	ND	5.29	8.40	1.01	ND	8.72	9.56	8.73
Cu	20.6	20.2	25.1	57.0	25.5	132	103	24.4
Hg	ND	ND	ND	ND	ND	ND	ND	ND
Mn	5.60	3.71	1.98	2.53	7.72	11.8	11.2	6.51
Ni	1.25	0.99	1.39	1.45	1.08	1.25	1.38	2.43
Pb	19.5	460	520	1.88	7.50	8.96	8.63	3.84
Sb	1.27	ND	ND	0.25	2.34	ND	ND	0.49
Sn	2.77	0.54	0.24	1.01	2.69	2.53	2.74	1.07
Tl	ND	ND	ND	ND	ND	0.10	ND	ND
V	1.37	0.70	0.47	0.86	2.22	2.28	2.36	1.14
Zn	24.1	19.1	15.2	2.86	29.2	24.3	33.4	4.36
PCDD/Fs	0.006	0.013	0.013	0.008	0.016	0.018	0.014	0.010
PCBs	0.008	0.074	0.097	0.079	0.045	0.030	0.025	0.020

ND: Not detected.
<1 km: NE1.
>1 km: NE2.
PCBs: Sum of the 7 environmental indicator congeners: PCB 28, 52, 101, 118, 153, 138, and 180.

Currently, EU directives establish maximum annual average air concentrations of 500, 20, 5, and 6 ng/m^3 for Pb, Ni, Cd and As (EC, 2004, 2008b). The current results indicate that all these four elements are clearly below the maximum established. Table 4 presents the PCDD/Fs and PCB air levels obtained in recent studies performed over the world. In general terms, air pollutant levels in the area under study were lower than most of the values reported in those recent studies.

3.3. Human health risks

Table 5 summarizes the average exposure for individuals living around the Mataró MSWI in 2011 and 2013. For metals, exposure

Table 4
Summary of some recent studies reporting levels of PCDD/Fs and PCBs in air samples, including the results of the current study.

PCDD/Fs (pg TEQ/m^3)	Type	Location	References
Levels around a MSWI			
0.011	Mean	Tarragona (Spain)	Vilavert et al. (2012)
0.007–0.042	Range	Tarragona (Spain)	Vilavert et al. (2014)
0.050	Mean	Taiwan	Chen et al. (2011)
0.060 (0.034–0.105)	Mean (range)	Taiwan	Wang et al. (2010)
0.028, 0.036, 0.039, 0.056	Seasonal means	Taiwan	Wang et al. (2013)
0.054 (0.014–0.103)	Mean (range)	China summer	Gao et al. (2014)
0.051 (0.020–0.090)	Mean (range)	China winter	Gao et al. (2014)
0.011 (0.008–0.015)	Mean (range)	Mataró (Spain 2008)	Rovira et al. (2010)
0.010 (0.006–0.013)	Mean (range)	Mataró (Spain 2011)	Present study
0.014 (0.010–0.018)	Mean (range)	Mataró (Spain 2013)	Present study
Other studies			
0.017–0.024	Range	Germany (2011 means)	Bruckmann et al. (2013)
0.01–0.19	Range	Brescia (Italy)	Colombo et al. (2013)
0.009–0.133	Range	Barcelona area (Spain)	Rovira et al. (2014)
PCBs (ng/m^3)	**Type**	**Location**	**References**
Levels around a MSWI			
0.034	Mean	Korea (Urban)	Heo et al. (2014)
0.036	Mean	Korea (Industrial)	Heo et al. (2014)
0.008–0.051	Range	Tarragona (Spain)	Vilavert et al. (2014)
0.026 (0.009–0.042)	Mean (range)	Mataró (Spain 2008)	Rovira et al. (2010)
0.064 (0.008–0.097)	mean (range)	Mataró (Spain 2011)	Present study
0.030 (0.020–0.045)	Mean (range)	Mataró (Spain 2013)	Present study
Other studies			
0.011 (0.004–0.027)	Mean (range)	China (Background)	Wu et al. (2011b)
0.093–8.566	Range	Brescia (Italy)	Colombo et al. (2013)
0.043 (0.027–0.98)	Mean (range)	Zurich (Switzerland)	Bogdal et al. (2014)

Table 5
Average exposure to metals (mg/(kg d)), PCDD/Fs (ng WHO-TEQ/(kg d)), and PCB (ng/(kg d)) of individuals living around the MSWI of Mataró in 2011–2013, and in the control point (B).

	Exposure 2011–2013				Average pathway contribution (%)		
	MSWI	<1 km	>1 km	B	Soil ingestion	Dermal absorption	Air inhalation
As	$3.05 \cdot 10^{-06}$	$3.40 \cdot 10^{-06}$	$4.28 \cdot 10^{-06}$	$2.86 \cdot 10^{-06}$	46	49	5
Cd	$1.11 \cdot 10^{-07}$	$1.60 \cdot 10^{-07}$	$2.07 \cdot 10^{-07}$	$9.63 \cdot 10^{-08}$	55	2	43
Co	$4.31 \cdot 10^{-06}$	$1.23 \cdot 10^{-05}$	$9.82 \cdot 10^{-06}$	$2.07 \cdot 10^{-05}$	74	26	0
Cr	$1.04 \cdot 10^{-05}$	$2.34 \cdot 10^{-05}$	$1.84 \cdot 10^{-05}$	$3.38 \cdot 10^{-05}$	65	23	12
Cu	$1.57 \cdot 10^{-05}$	$5.23 \cdot 10^{-05}$	$4.56 \cdot 10^{-05}$	$2.91 \cdot 10^{-05}$	45	16	39
Mn	$2.65 \cdot 10^{-04}$	$7.54 \cdot 10^{-04}$	$7.46 \cdot 10^{-04}$	$1.52 \cdot 10^{-03}$	74	26	0
Ni	$4.22 \cdot 10^{-06}$	$6.85 \cdot 10^{-06}$	$5.33 \cdot 10^{-06}$	$8.88 \cdot 10^{-06}$	69	25	6
Pb	$1.25 \cdot 10^{-05}$	$8.65 \cdot 10^{-05}$	$1.03 \cdot 10^{-04}$	$2.38 \cdot 10^{-05}$	41	15	44
Sb	$4.95 \cdot 10^{-07}$	NC	NC	$6.85 \cdot 10^{-08}$	NC	NC	100
Sn	$1.48 \cdot 10^{-06}$	$1.85 \cdot 10^{-06}$	$1.97 \cdot 10^{-06}$	$3.15 \cdot 10^{-06}$	63	22	15
Tl	$1.44 \cdot 10^{-07}$	$3.88 \cdot 10^{-07}$	$3.56 \cdot 10^{-07}$	$6.29 \cdot 10^{-07}$	74	26	NC
V	$3.09 \cdot 10^{-05}$	$1.22 \cdot 10^{-04}$	$8.85 \cdot 10^{-05}$	$2.00 \cdot 10^{-04}$	73	26	1
Zn	$4.91 \cdot 10^{-05}$	$1.31 \cdot 10^{-04}$	$1.19 \cdot 10^{-04}$	$1.79 \cdot 10^{-04}$	69	25	6
PCDD/Fs	$3.50 \cdot 10^{-06}$	$5.36 \cdot 10^{-06}$	$4.69 \cdot 10^{-06}$	$2.87 \cdot 10^{-06}$	9	9	82
PCBs	$1.10 \cdot 10^{-02}$	$3.49 \cdot 10^{-02}$	$2.80 \cdot 10^{-02}$	$1.72 \cdot 10^{-02}$	7	32	61

NC: Not calculated because environmental concentration was below detection limit.

ranged between $9.63 \cdot 10^{-8}$ mg/(kg day) for Cd in the background area, and $7.54 \cdot 10^{-4}$ mg/(kg day) for Mn at <1 km of the facility. Regarding PCDD/Fs and PCBs, the human exposure ranged between $2.87 \cdot 10^{-6}$ and $5.36 \cdot 10^{-6}$ ng WHO-TEQ/(kg day), and between $1.10 \cdot 10^{-2}$ and $3.49 \cdot 10^{-2}$ ng/(kg day), respectively. In general terms, the main exposure pathway for metals was soil ingestion, with the exceptions of As, for which it was dermal absorption, for Pb, due the high air levels found in 2011 in Mataró (<1 km and >1 km), and for Sb, which was only detected in air samples. The main exposure pathway of PCDD/Fs and PCBs was air inhalation, with a contribution percentage of 82% and 61%, respectively, to total exposure. Based on the results of recent studies, in which the dietary intake of pollutants by the population in Catalonia (Martorell et al., 2011; Perelló et al., 2012, 2014) was estimated, dietary exposure of As, Cd, Hg, Pb, PCDD/Fs and PCBs would still mean between 93% and 99% of the total (dietary + environmental) exposure to these pollutants. Furthermore, and taking into account the PCDD/F intake from dairy products calculated from measured deposition in an Alpine region of Italy (Schiavon et al., 2013), the environmental exposure to PCDD/Fs found in Mataró is >99% of total exposure. Results of exposure assessment here presented are in accordance with a biomonitoring study (Parera et al., 2013) carried out in Mataró that found similar PCDD/F and PCB blood levels in population regardless distance to MSWI.

Fig. 2 depicts the carcinogenic risks associated to the exposure to carcinogenic elements, PCDD/Fs and PCBs around Mataró MSWI, according to the environmental levels found in the 2011 and 2013 campaigns. Only for Cr inhalation, the maximum admissible carcinogenic risk level set by the Spanish legislation (10^{-5}) (Ministerio de Medio Ambiente, 2007) was exceeded. However, the US EPA considers as acceptable values in a range between 10^{-6} and 10^{-4}, taking into account the variability, the uncertainty related to the individual characteristics of each subject, and the risk accumulation of the co-exposure to different substances (US EPA, 1996). In the present study, only total Cr levels were analyzed, while toxicological parameters for hexavalent chromium were used. For that reason, Cr (VI) levels in air and soil were assumed to be 1/6 of the total Cr concentration (US EPA, 1998). Similarly, As was assumed to be exclusively in its inorganic form for risk assessment. The assumptions for As and for Cr have been supported by results from recent studies (Brown et al., 2014; Huang et al., 2014).

With respect to non-carcinogenic risks, all metals, PCDD/Fs and PCBs showed a hazardous quotient (HQ) notably below the unity,

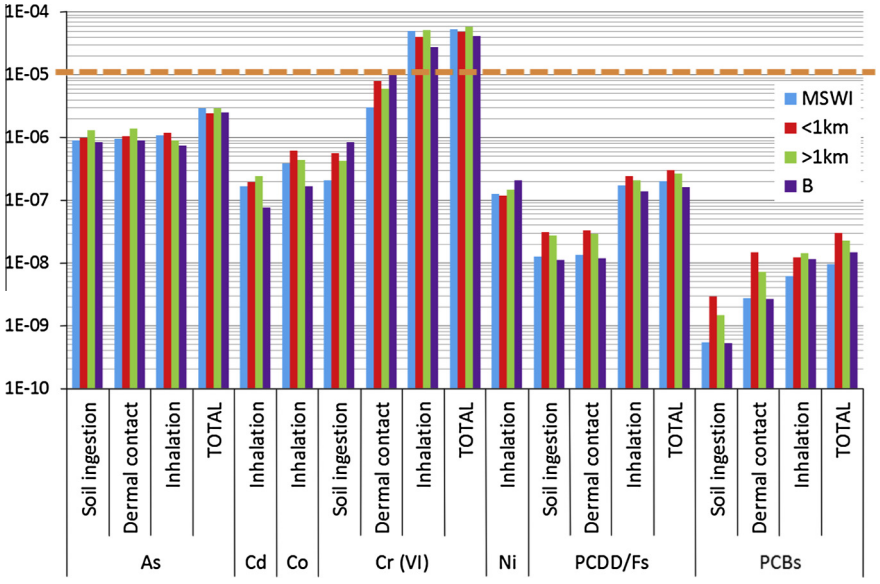

Fig. 2. Average carcinogenic risks inside and around the MSWI according to the results of the 2011 and 2013 sampling campaigns.

which is considered the threshold value (HQ = 1). The HQs for exposure to PCDD/Fs and PCBs were clearly lower than 0.1 (HQ < 0.1) in all sampling points, in both campaigns. For metals, none of the analyzed elements exceeded an HQ value of 0.2, with the only exception of Mn, which reached a HQ of 0.4.

In the present study, updated information regarding the environmental concentrations of metals, PCDD/F and PCBs in the vicinity of the MSWI of Mataró (Catalonia, Spain) was presented. Temporal trends of pollutant levels after the implementation of several technical improvements in the facility, were evaluated. No significant changes were observed in the concentrations of chlorinated compounds (PCDD/Fs and PCBs) in either soil or air. Similarly, metal concentrations in soil did not vary significantly with time, while the levels of a number of elements were significantly higher in the background survey (2008) than in the most recent studies (2011 and 2013). In conclusion, based on the above and still pending to acquire new data, it seems that the exposure to metals, PCDD/Fs and PCBs does not mean additional non-carcinogenic risks for the population living in the neighborhood of the Mataró MSWI. Both, carcinogenic and non-carcinogenic risks due the exposure to the environmental pollutants here analyzed were similar, or even lower, than those found in the previous (2008) sampling campaign, being also acceptable according to national and international standards. Studies such as the present one, are essential to detect any possible changes in the environmental levels of the pollutants emitted by MSWIs, due to potential variations in the stack emissions. They are also a key step to assess additional health risks for the population living in the surroundings of MSWIs.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.wasman.2015.05.039.

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